Changes in nitrogen cycling during the past century in a northern hardwood forest

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Nitrogen (N) availability, defined here as the supply of N to terrestrial plants and soil microorganisms relative to their N demands, limits the productivity of many temperate zone forests and in part determines ecosystem carbon (C) content. Despite multidecadal monitoring of N in streams, the long-term record of N availability in forests of the northeastern United States is largely unknown. Therefore, although these forests have been receiving anthropogenic N deposition for the past few decades, it is still uncertain whether terrestrial N availability has changed during this time and, subsequently, whether forest ecosystems have responded to increased N deposition. Here, we used stable N isotopes in tree rings and lake sediments to demonstrate that N availability in a northeastern forest has declined over the past 75 years, likely because of ecosystem recovery from Euro-American land use. Forest N availability has only recently returned to levels forecast from presettlement trajectories, rendering the trajectory of future forest N cycling uncertain. Our results suggest that chronic disturbances caused by humans, especially logging and agriculture, are major drivers of terrestrial N cycling in forest ecosystems today, even a century after cessation.

^{15}N | land-use history | Mirror Lake | nitrogen availability | paleoecology

Humans have increased the amount of reactive N on earth (1), leading to negative environmental consequences that include reduced forest growth and eutrophication of surface waters (2). In contrast to Europe, where anthropogenic N deposition is generally greater, the consequences in northeastern North American forests are less clear because there has been little obvious evidence of forest eutrophication. Although N is a key regulator of ecosystem processes (3, 4), it is unknown whether N availability in North American temperate forests has been increasing or decreasing during the past few decades.

Theoretically, N availability should have been increasing over the past few decades because forest biomass accumulation slows with stand age (5), base cations become depleted because of acid rain-enhanced leaching (6, 7), and industrially produced N is deposited on forests from the atmosphere (8). Alternatively, elevated levels of atmospheric CO_2 could be reducing N availability by stimulating microbial immobilization of N (9, 10). To date, the only estimates of historical changes in terrestrial N availability are derived from changes in streamwater nitrate concentrations in forested catchments that suggest that forest N availability has declined in many parts of the northeastern United States since the 1970s (11, 12). Unfortunately, streamwater nitrate concentrations ambiguously reflect terrestrial N availability because they can be influenced by in-stream chemical processing (13, 14). Also, streamwater nitrate records are generally restricted to the past few decades. Thus, these records cannot provide information about N availability before elevated N deposition, determine whether Euro-American settlement two centuries ago altered N availability, or establish baseline N availability before Euro-American settlement.

To begin to reconstruct past terrestrial N availability, we measured stable N isotopes in tree rings and lake sediments for a forest typical of the northeastern United States. Our analysis focused on the catchment of Mirror Lake, a 15-hectare oligotrophic lake in the White Mountains of New Hampshire. This site receives anthropogenic N in precipitation typical of the region (15), N flux in streams has been monitored since 1980, and its well-documented history shows land use similar to that in much of the northeastern United States (16). Of particular interest, the majority of the 103 hectares of forested watershed was cleared for agriculture starting in 1790, and forest regrowth commenced in the first two decades of the 20th century.

Although the N cycle is complicated relative to other biologically important elements, recent analytical and conceptual advances have made it possible to measure and interpret the natural abundance of stable N isotopes in terrestrial ecosystems. Because N is supplied to and consumed by plants and microbes on a variety of temporal and spatial scales, the concept of terrestrial N availability has been gradually refined (17). Ecosystems with high N availability, such as those that are fertilized with N, exhibit high values of natural abundance ^{15}N:^{14}N ratios (δ^{15}N) in leaf tissue and soil (18). As with tree leaves (19), enrichment of wood with ^{15}N occurs with high N availability, and therefore dendroisotopic records have been used to reconstruct past N availability (20, 21).

By combining three independent but temporally synchronous estimates of N availability to forests (tree rings, lake sediments, and streamwater nitrate data), we reconstructed terrestrial N availability for the past millennium. Increment bores were collected from 22 trees distributed among the three subwatersheds draining into Mirror Lake following standard procedures. Patterns of δ^{15}N were determined by using isotope ratio mass spectrometry. Because such dendroisotopic records are limited to approximately the past century in this region because of forest clearance, we extended estimates of N availability to ≈900 A.D. after first matching N isotope records from a sediment core from Mirror Lake with contemporaneous tree-ring records. We then derived a baseline for N availability from the sediment record before Euro-American settlement to estimate presettlement N availability and to quantify the degree of forest recovery [see supporting information (SI) *Methods*].

Results and Discussion

Dendroisotopic analyses revealed that N availability in the forests surrounding Mirror Lake has been declining steadily since cessation of agriculture and forestry 75 years ago despite

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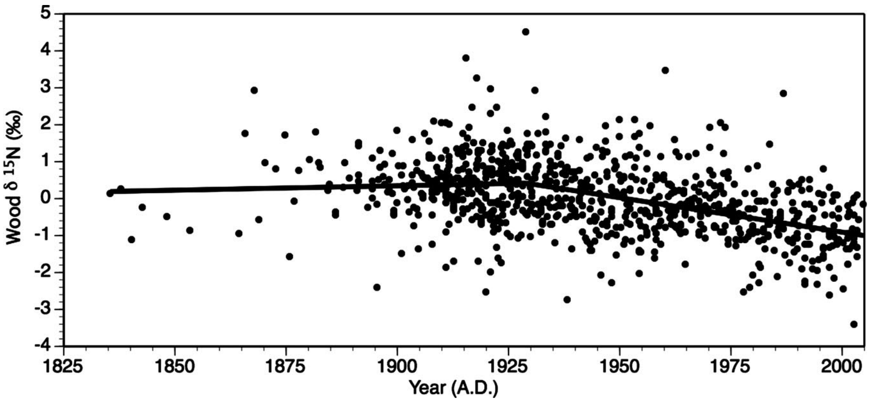

Fig. 1. History of terrestrial N availability from tree rings at Mirror Lake. δ^{15}N in 857 wood segments from 22 trees in the Mirror Lake watershed, with a piecewise linear regression line. Each tree was standardized to a mean δ^{15}N of 0‰. Partition analysis indicates that the inflection point is at 1929 (95% confidence interval: 1914 to 1937).

increasing anthropogenic N deposition during some of this time (Fig. 1). Across all trees sampled, wood δ^{15}N declined at a rate of 0.019 ± 0.001‰ per year since 1929, an inflection point determined with piecewise linear regression (22). Best estimates of the onset of decline in δ^{15}N range from 1914 to 1937 (95% confidence interval) with all dates in this interval producing similar estimates of the total decline in wood δ^{15}N from the onset to the present. Declines in wood δ^{15}N were consistent among individual trees (SI Fig. 4) but were not associated with changes in wood N concentrations (SI Fig. 5) nor changes in forest composition (SI Fig. 6). Instead, rates of decline of wood δ^{15}N matched 20 years of declining nitrate flux from inlet streams to Mirror Lake (SI Fig. 7), suggesting that N availability to forests has been declining steadily during the latter part of the 20th century.

Analyses of N isotopes in lake sediments further indicate the decline in forest N availability over the past 75 years. From 1929 to 2005, reductions in wood δ^{15}N occurred synchronously with declines in sediment δ^{15}N, indicating that sediment δ^{15}N primarily reflects terrestrial N availability (Fig. 2*A*). Our interpretation is based on observations that high terrestrial N availability leads to high δ^{15}N in lake sediments as ^{15}N-enriched organic matter and nitrate enter the lake (23, 24). The magnitude of δ^{15}N reduction in sediments since 1929 (0.9‰) was slightly less than that observed in tree rings (1.4‰), but this result is expected because atmospheric N inputs to the lake dilute terrestrial N. Agreement among the tree ring, lake sediment, and streamwater nitrate data shows a pervasive pattern of reduced N availability in forests during the past several decades.

We measured several other geochemical characteristics in sediment from Mirror Lake, including C and N concentrations,

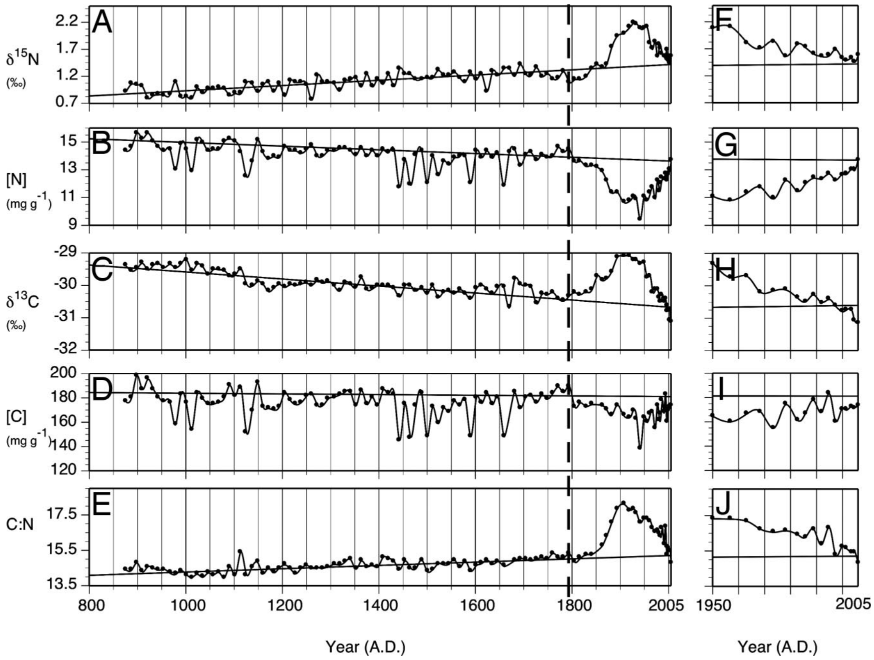

Fig. 2. Geochemical data from surface sediment core of Mirror Lake. Linear regression lines are shown for δ^{15}N (*A* and *F*), δ^{13}C (*C* and *H*), and C:N (*E* and *J*). Seventy-five percent quantile regression for [N] (*B* and *G*) and [C] (*D* and *I*) was used to minimize the influence of several sediment samples unusually high in mineral material, similar to one that was deposited immediately after a powerful hurricane in 1938. All regressions were performed on samples deposited earlier than 1800. The most recent samples, from 1950 to 2005, are shown in *F*–*J*. The vertical dashed line indicates the first record of Euro-American settlement in the Hubbard Brook Valley in 1790.

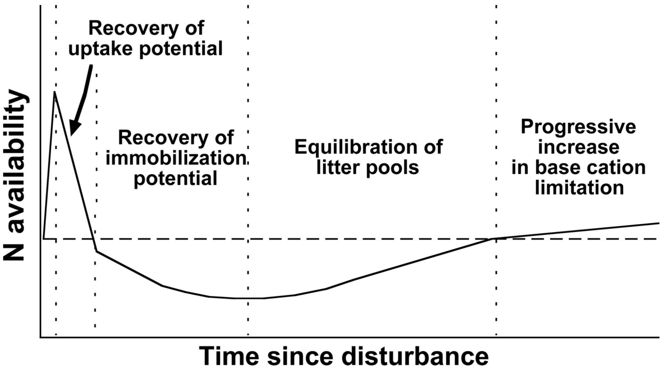

Fig. 3. Conceptual diagram of N availability for northern hardwood forests during secondary succession. N availability rises with disturbance, but the recovery of vegetation uptake potential generally decreases N availability soon after. N availability can continue to decline, potentially even below predisturbance N availability, if soil and litter fractions were depleted by disturbance and their recovery immobilizes N. The recovery of immobilization might be followed by an increase in N availability as these pools equilibrate, but there has been little research on these long-term dynamics. Over longer time scales, N availability increases with ecosystem development and increasing base cation limitation. Axes are not to scale.

C:N, and δ^{13}C. Based on standard conventions in paleolimnology, since 1928, lake primary productivity and erosion inputs have declined in Mirror Lake (Fig. 2 *B–D*). The concentrations of C and N in bulk sediments, because they reflect the proportion of inorganic and organic material, provide an index of the relative amount of inorganic material eroded from the catchment because of disturbance or other processes (25). Since 1928, C and N concentrations have been declining in Mirror Lake, indicating decreased inorganic inputs (Fig. 2 *B* and *D*). Sediment C:N scales positively with the quantity of terrestrial organic matter transported to the lake. Autochthonous material such as algae has a C:N $<$10:1, and terrestrial plant material measures $\approx$20–70:1 (26). The C:N has been declining in Mirror Lake sediments for the past several decades (Fig. 2*E*). Sediment δ^{13}C is positively correlated with lake productivity, because algal photosynthetic demand for C is the dominant fractionation pathway (27), and it has been declining for the past several decades (Fig. 2*C*).

Declining N availability over the past 75 years is likely associated with forest recovery from the previous 150 years of land use. Beginning in 1790, Euro-American activities such as clearing forest vegetation and tilling the soil would have reduced plant uptake of N and fixation of C (28). These activities increased the relative availability of N in terrestrial ecosystems surrounding Mirror Lake as recorded by the sharp increase in sediment δ^{15}N until 1928. Forest clearance was recorded in the sediment record as increased pollen from herbaceous taxa and decreased tree pollen (SI Fig. 6), dilution of organic matter by mineral inputs eroded from the catchment, and increased sedimentary C:N ratios from those characteristic of algal production toward those characteristic of terrestrial plant material (Fig. 2*E*).

The prolonged decline in N availability in the Mirror Lake watershed during the past 75 years is likely due to several autogenic processes that follow cessation of chronic disturbance from agriculture and logging (Fig. 3). Clearance of forest and subsequent agriculture result in loss of forest soils, decline in soil C:N, and reduction of N immobilization and increased N availability. After clearance, recovery of plant N uptake in strongly aggrading forests causes immediate declines in N availability, but this process does not continue to cause declines much more than 5 years after a clear cut (29). The main mechanisms causing declining N availability on a multidecadal timescale are: (*i*) recovery of N immobilization potential via C additions, (*ii*) increasing soil C:N after loss or degradation of the forest floor, (*iii*) soil organic matter accumulation (29, 30), and (*iv*) the production of coarse woody debris (31).

Our paleoecological analysis of sediments suggests that N availability in northeastern forests has only recently reached baseline levels forecast from millennium-long sedimentary analyses (Fig. 2 *F–J*). Before 1800, baseline sediment δ^{15}N had been increasing at a rate of 0.05‰ per century, consistent with theories of ecosystem development such as reductions of base cation availability (6, 32) and the forested ecosystem as a steady state mosaic that can include slow unidirectional change (29). At this rate, sediment δ^{15}N would have been $\approx$1.4‰ in 2005 had the ecosystems not been chronically disturbed. Indeed, sediment δ^{15}N in the most recently deposited sediments is only just reaching the projected modern baseline level, after being 0.8‰ greater than baseline as recently as 1927. Similar analyses of baseline trajectories for sedimentary C and N concentrations, C:N ratios, and δ^{13}C signatures also indicate that Mirror Lake and its surrounding forests are just now reaching baseline levels (Fig. 2 *G–J*).

Declines in terrestrial N availability reconstructed from sedimentary and dendrochronological records corroborate reports of declining nitrate export from northeastern United States forests over the past 30 years. Although it has been suggested that N availability should have been increasing in these forests as a result of elevated atmospheric N deposition (33, 34), apparently the increasing N immobilization potential of the forests surrounding Mirror Lake has sufficiently overwhelmed the $\approx$1 $g \cdot m^{-2} \cdot y^{-1}$ of anthropogenic N that has been deposited since 1970 (35). To date, N deposition might have slowed the rate of decline of N availability as immobilization potential has recovered since 1929, but it has not led to absolute increases in N availability. Our results show that chronic disturbances, especially logging and agriculture, have determined the trajectory of terrestrial N availability over the past century, but N availability in the Mirror Lake watershed is now approaching the level forecast by presettlement trajectories. Although the future contribution of the recovery of immobilization potential is uncertain, as the effects of past land use diminish, further monitoring and research will be necessary to determine the relative contribution of other factors, such as N deposition, elevated CO_2, and altered temperatures, to the future trajectory of N availability in the forests of the region.

Methods

Site Description. Mirror Lake is a small (15 hectare), oligotrophic lake in the White Mountains (16). The surrounding vegetation is northern hardwood forest containing deciduous and coniferous tree species. The first Euro-American settlers in the watershed were recorded in the 1790 United States Census, and soil tillage for row-crop agriculture, grazing by sheep in pastures, and selective logging all occurred subsequently. Nitrate was deposited on Mirror Lake in bulk wet deposition from 1964 to 2003 at a rate of 0.48 g $N \cdot m^{-2} \cdot y^{-1}$, primarily from industrial sources. Some dry deposition of nitrate and ammonium does occur, primarily from vehicular sources (36). From 1989 to 2004, the average amount of dry N deposition was $\approx$0.05 $g \cdot m^{-2} \cdot y^{-1}$, 8% of the total N deposition.

Tree Cores. Increment bores were acquired from 22 trees of five different species divided among the three subwatersheds of Mirror Lake. A 5.15-mm diameter increment bore was acquired from each tree at $\approx$1 m above the forest floor. Seventy-five percent of trees that we sampled were established after 1890, and none of the sampled trees were established after 1921. The trees we selected randomly were therefore representative of the age structure of trees in the watershed. Although the precise locations of former land use are unknown, our sampling strategy maximized spatial coverage of the watershed and therefore might have included some trees in locations that had not been farmed or logged.

After determination of ring widths, each bore was cut into 30-mg sections with sections divided along boundaries between ring widths. On average, a section contained wood deposited during 2.75 years. The ^{15}N:^{14}N ratio of each section was determined at the University of California, Davis, on a PDZ Europa 20-20 isotope ratio mass spectrometer fitted with sequential traps of $MgClO_4$, NaOH on solid support (Carbosorb, Sydney, Australia), and a cold trap in liquid N_2. Analytical error (one standard deviation) was 0.3‰ for these samples (37). Although the choice of tree species from which to obtain increment bores might affect wood N dynamics because of differences in sapwood extent, formation of secondary compounds, and radial permeability, patterns of ^{15}N in tree rings largely reflect the chronology of N availability for the tree (20). There is little evidence for remobilization of N out of old wood and deposition of newly acquired N, if at all significant, has little effect beyond the previous 10 years (38). With the δ^{15}N signature of modern atmospheric N deposition near zero, it is unlikely that declines in wood δ^{15}N before the onset of industrial N pollution could be explained by changes in the ^{15}N signatures of deposited N.

To determine the pattern of wood δ^{15}N over time, we standardized δ^{15}N data for each bore by subtracting the mean δ^{15}N of the bore from each point so that each bore has a mean δ^{15}N of 0‰. This removes patterns that might arise as a result of differences in ages among trees. The data were then subjected to piecewise linear regression using the nonlinear model algorithm of JMP 5.01 (SAS Institute, Cary, NC). The inflection point was adjusted manually by single-year increments, and then the model with the lowest sum of squares was used to select the best inflection point. Adding an additional inflection point after 1929 did not lead to a significant change in the rate of δ^{15}N decline over time. We also performed simple linear regressions of δ^{15}N against section age since 1929 or for the entire record for each increment bore (SI Fig. 4).

Sediment Core. We obtained a 102-cm long sediment core from Mirror Lake on August, 3, 2005, using a hand-driven 7-cm diameter polycarbonate tube fitted with a piston. The sediment was sectioned into 0.5-cm intervals from 0 to 10 cm depth and into 1-cm intervals from 10 to 102 cm depth. A portion of the sediment from each interval was weighed, dried in an oven at 65°C until no further mass loss was observed, and ground with a mortar and pestle. The chronology of the sediment core was established with ^{210}Pb dating at the Saint Croix Watershed Research Station in Minnesota. ^{210}Pb was measured at 21 depth intervals by α spectrometry, and dates and sedimentation rates were determined according to the constant rate of supply model (39). Sedimentation rates were constant between 12 and 18 cm, so we used the average sedimentation rate from these intervals, 11.3 year·cm^{-1}, to calculate ages for samples below 18 cm, or before 1829 (SI Fig. 8). This age-depth model, based on extrapolation of sedimentation rates at levels deeper than detectable ^{210}Pb activity, is similar to those in previous studies of Mirror Lake sediments (40). Elemental and isotopic analysis for C and N were conducted at the Stable Isotope Lab at the University of Regina by using standard methods on a Thermoquest (Finnigan-MAT, San Jose, CA) Delta Plus mass spectrometer interfaced with a Carlo Erba (Carlo Erba, Milan, Italy) NC2500 elemental analyzer. Analytical error was <0.1‰ for δ^{15}N and 0.2‰ for δ^{13}C.

Nitrogen fixation in Mirror Lake is minimal (16). With all else equal, lake productivity does not affect sediment δ^{15}N. There is neither evidence that denitrification occurs in the water column nor that the patterns in δ^{15}N seen in the sediment record could be caused by denitrification in the sediment column. Dissolved organic N concentration is very low in streams draining the Mirror Lake watershed, and changes in organic and inorganic N transport are unlikely to determine patterns in sediment δ^{15}N. Previous work on Mirror Lake sediments has revealed changes in sedimentation rates and patterns over time, and thus C and N concentrations are not strictly proportional to absolute erosion rates within the catchment (16). Although sediment δ^{15}N is positively associated with the input of ^{15}N-enriched material from terrestrial ecosystems with high N availability, it is uncertain to what degree variation in sediment δ^{15}N is also influenced by variation in inputs from pools that have different δ^{15}N, such as mineral soil, forest floor, or leaves.

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